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October 1981

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PAD FACILITY FOR THE CALIBRATION OF GAMMA-RAY MEASUREMENTS ON ROCKS

L. Løvborg, E. M. Christiansen,
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Abstract. A series of six concrete pads that simulate rock outcrops of known radioactive concentrations have been installed at Risø National Laboratory. The facility is intended for the calibration of portable counters and spectrometers and is accessible to uranium geologists in Europe and elsewhere. All of the pads are 3 m in diameter and 50 cm thick and contain uranium, thorium, and potassium in various proportions. The concentrations of eU, Th, and K with associated standard errors have been determined from the combined use of laboratory assays and radiation monitoring data. The evaluations of instrumental sensitivity factors and stripping ratios are performed with weighted least-squares techniques. Sensitivity factors of gross-counting field scintillometers are supplied in units of radioelement concentration (Ur). In addition to being especially suited to geologic calibrations, the facility may be applied as a source of gamma-ray exposure rates for calibrating environmental survey instruments.

INIS-descriptors: CALIBRATION; CONCRETES; DOSE RATES; ENVIRONMENT; EXPLORATION; GAMMA SPECTROMETERS; GEOLOGIC DEPOSITS; POTASSIUM; RADIOACTIVITY; ROCKS; SCINTILLATION COUNTERS; SENSITIVITY; SIMULATION; THORIUM; URANIUM

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October 1981
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1. INTRODUCTION

This report is addressed to geologists who are involved in gamma-ray assays of radioelement concentrations in outcropping rock and exposed radioactive mineralization. Measurements like these are of great importance in uranium exploration and may be additionally applied as an aid to geological mapping. The proper determination of the associated instrument calibration factors is conveniently performed on specially constructed concrete pads with enhanced and known radioelement concentrations. The pad facility described here consists of six single pads which are 3 m in diameter and 50 cm thick and are loaded with U, Th, and K in various proportions. The facility is located at Risø National Laboratory and may serve calibration tasks implied by uranium exploration in the European countries.

In the following the customers of instrument calibration at Risø are supplied with up-to-date information on the pads and the way they are used. A similar description by Løvborg et al. (1978) is now obsolete and should no longer be consulted. The new set of specifications is based on the recent addition of two extra pads to the original series of only four pads. The enlargement of the facility was accompanied by laboratory assays and radiation monitoring for determining the radioelement concentrations in the pads with the greatest possible accuracy. A further improvement consists in the development of efficient computer programs for processing calibration counts recorded on the pads. Instrumental sensivity factors and stripping ratios are henceforth evaluated using least squares techniques in which the known variances of the pad concentrations are taken into consideration.

The calibration pads at Risø simulate rock outcrops with areas of about 7 m^2. The service offered to users of the facility includes an estimate of counter and spectrometer sensitivities

in field assays of much larger exposed targets. This optional step in the calibration procedure is based on gamma-ray transport calculations in cylindrical geometry. Furthermore it is possible to provide a total-count calibration in units of μR/h based on the experimental gamma-ray exposure rates contributed by the individual radioelement concentrations in the pads.

2. DESCRIPTION OF THE FACILITY

2.1. Location and layout

Risø National Laboratory is the largest research institution in Denmark. It is located on the A6 main highway between Roskilde and Hillerød, 6 km north of the former. Figure 1 indicates

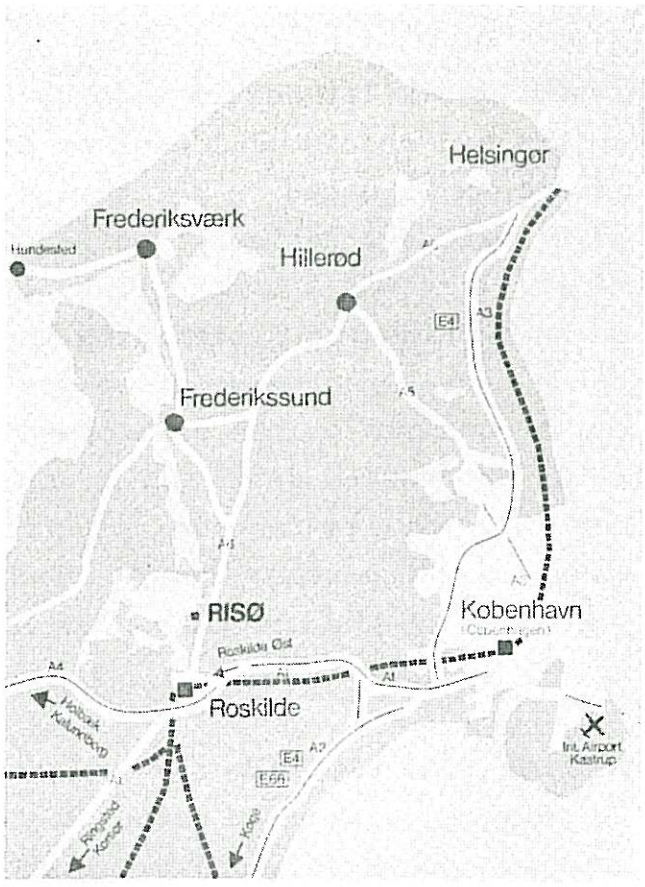

Fig. 1. Location of Risø National Laboratory north of Roskilde, Denmark.

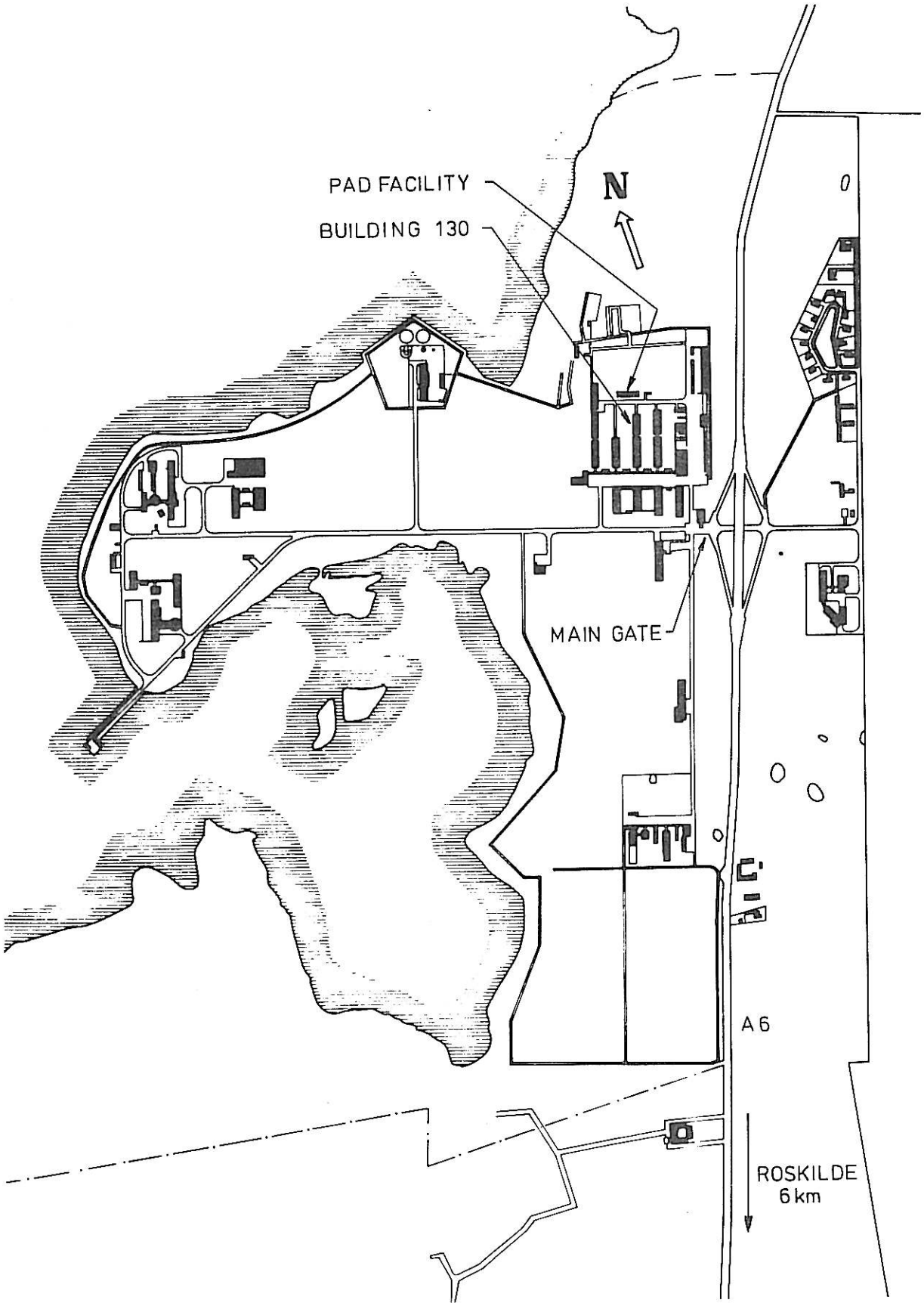

Fig. 2. Sketch of the Risø area showing pad facility and domicile of Nuclear Geophysics Group in Building 130.

the connections to Copenhagen and the International Airport at Kastrup. The calibration service is operated by the Nuclear Geophysics Group which has its laboratory and offices in Building 130 on the right-hand side of the main gate (see Fig. 2). The pads occupy a strip of land adjacent to the access road north of the building.

A plan of the facility is presented in Fig. 3 which also shows the designations used for the pads. K-1, K-2, T-1, and U-1 are the original series of pads. These possess essentially pure concentrations of a single radioelement: K, Th, or U. They date from 1971 and were previously numbered from 0 to 3. UT-1 and UT-2 are the new pads which were constructed in 1979 and are

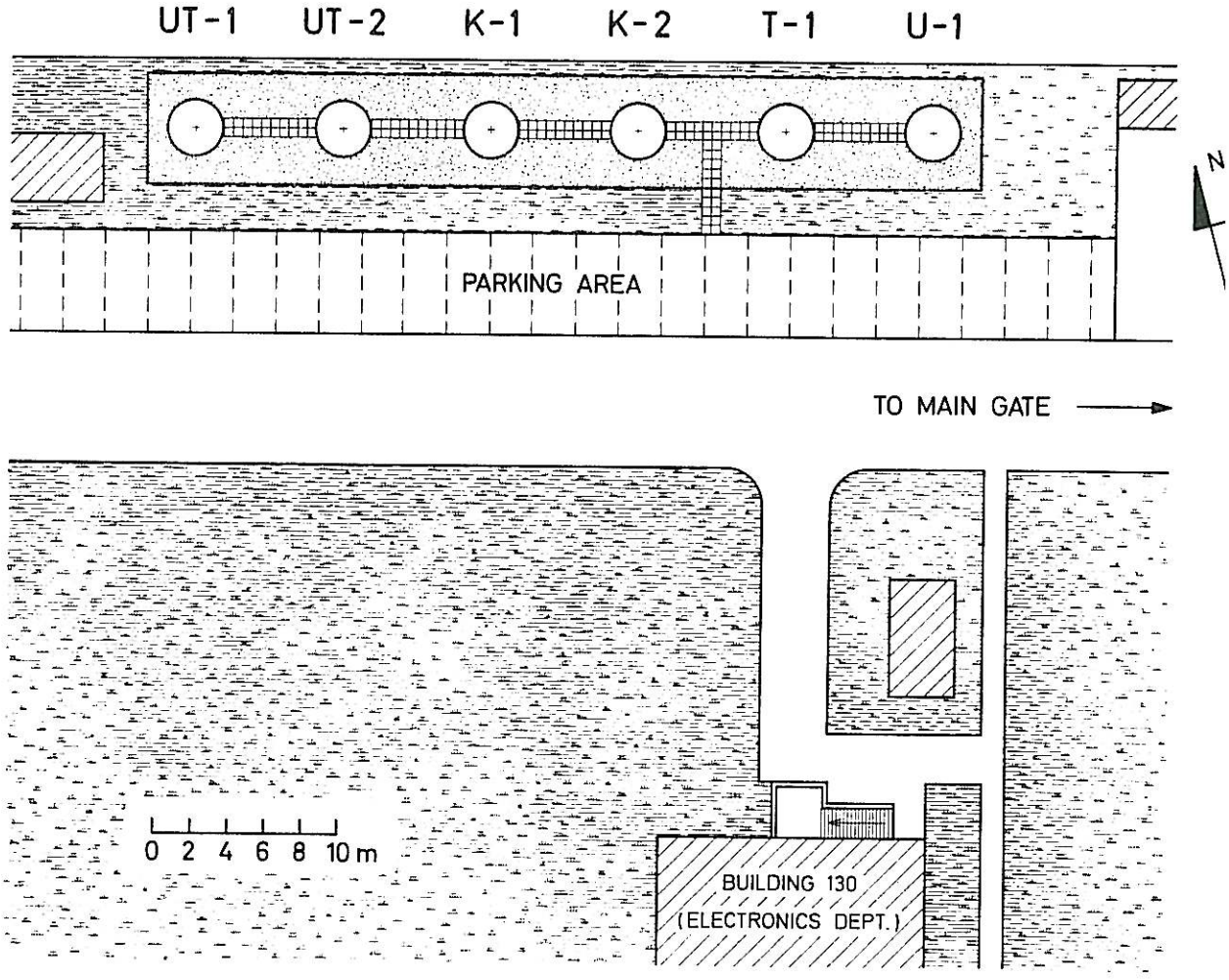

Fig. 3. The arrangement of the six calibration pads.

characterized by mixed contents of U and Th. In Fig. 4 the facility is viewed from the eastern end with survey instrumentation positioned on U-1 and K-1. The pad cylinders are embedded in gravel so that only their top faces are visible. Since there is no shelter on the pads, instrument calibration is impracticable on days with snowfall or heavy rain. Between December and March the pads may be periodically blocked by snow.

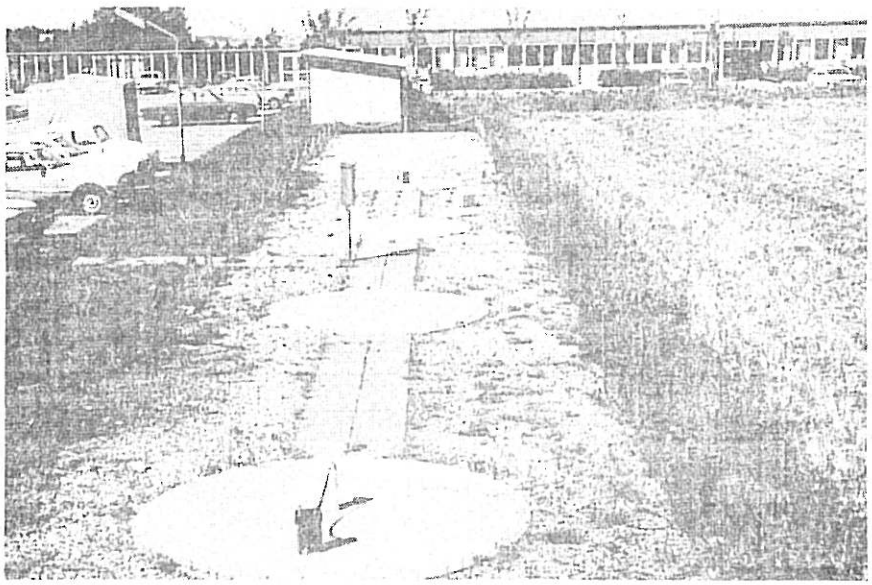

Fig. 4. View of the facility from the east.

2.2. Constructional details

The pads are made of concrete that has been prepared from mixtures of 65 to 70% aggregate material, 20 to 25% cement, and less than 10% water. Beach sand has been used as the predominant aggregate material for all pads except K-2 which has an aggregate of Norwegian potassium feldspar with 10% K. The radioactive admixtures for pads enriched in U and Th are specified in Table 1. K-1 is the only pad that does not carry a load of material with high radioelement content. It consists of plain concrete and has a small natural concentration of 1% K.

The four pads dating from 1971 were manufactured on the spot (see Fig. 5). The mixtures for the two most recently made pads

Table 1. Radioactive admixtures for pads enriched in U and Th.

Pad	Admixture				
	Kind	Place of origin	% U	% Th	Load (kg)
U-1	Uranium ore	Saskatchewan, Canada	10	-	18
T-1	Monazite sand	Malaysia	-	6	18
UT-1	Peralkaline nepheline syenite	Kvanefjeld, S.W. Greenland	0.07	0.2	540
UT-2	Peralkaline nepheline syenite	Kvanefjeld S.W. Greenland	0.07	0.2	180

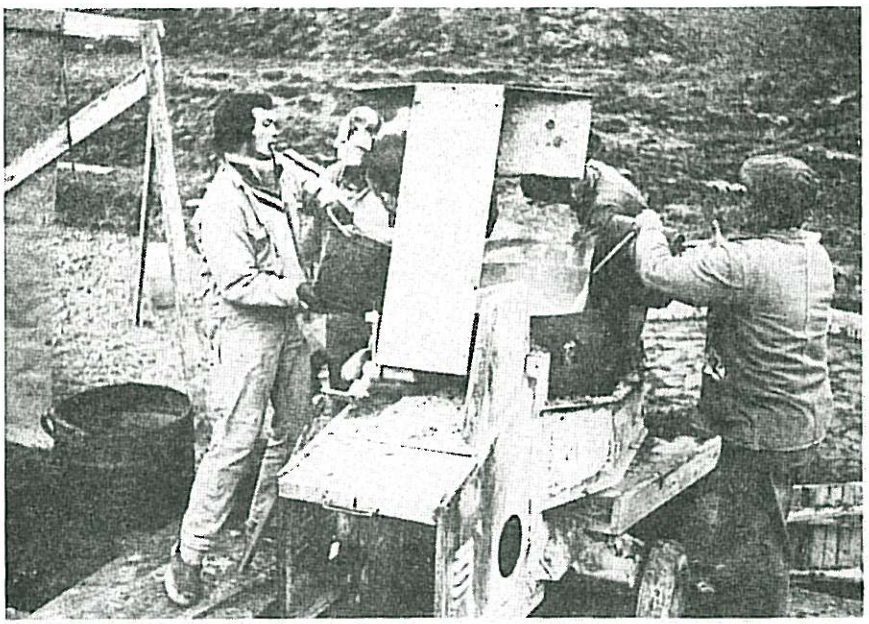

Fig. 5. Pad manufacture in 1971.

were prepared at a concrete factory in Roskilde and arrived at Risø ready for use (Figs. 6 and 7). In both cases the concrete raw materials were blended for a prolonged period before water was added. The uranium ore for U-1 as well as the nepheline syenite for UT-1 and UT-2 were supplied as fine powders. These

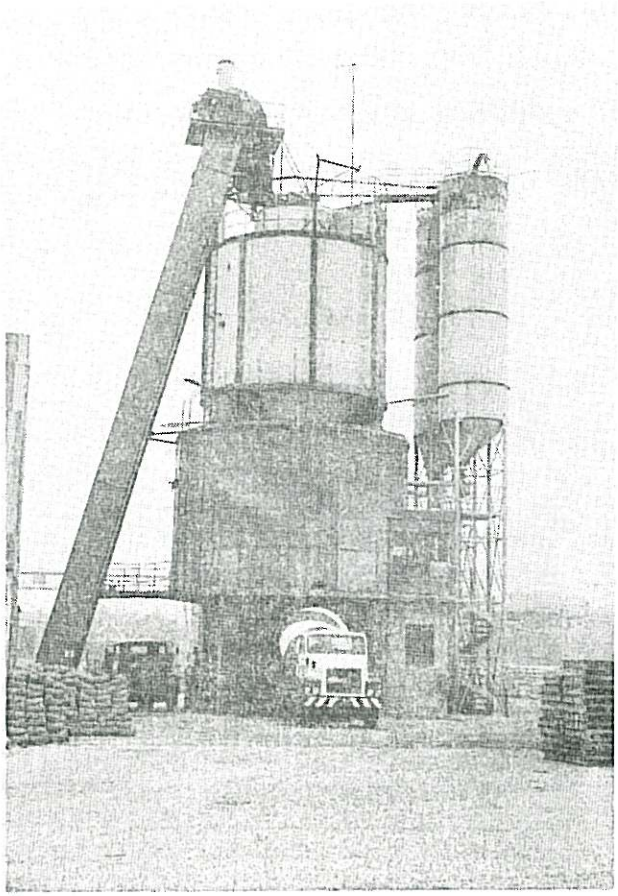

Fig. 6. Pad manufacture in 1979. Truck leaving factory with ready-made concrete mixture.

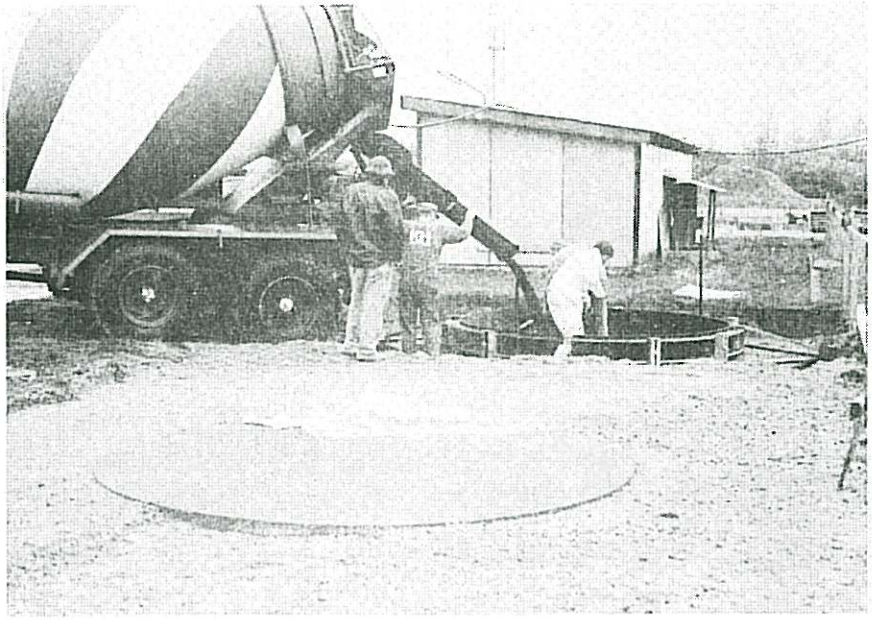

Fig. 7. Pouring of mixture into re-usable form.

were pre-mixed with pure cement. The construction of each pad was aided by a wooden form in which portions of the mixtures were alternately placed and rammed. Figure 8 shows a nearly completed pad. An example of solidified pad with detached form compartments is presented in Fig. 9. All pads have slightly convex surfaces that permit rainwater to run off into drain

Fig. 8. Ramming of wet mixture.

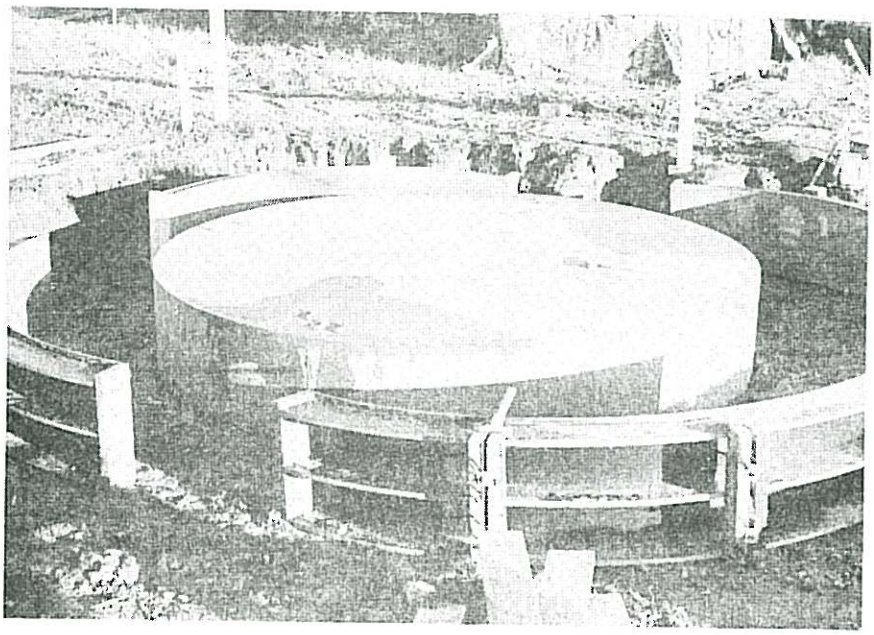

Fig. 9. Solidified pad.

pipes in the gravel filled in at the site. The pads are spaced at intervals of 8 m and are connected by a line of walking stones. Users of the facility may be supplied with 220/380 V, 50 Hz electrical power from a service frame erected between K-2 and T-1.

2.3. Chemical and physical characteristics of the pads

Table 2 shows the chemical compositions of the pads. It is not surprising that the pads exhibit a chemical similarity with calcarous sandstone. An exception is the K-2 with its high content of potassium feldspar. Table 3 summarizes relevant data that are more or less common to the pads, i.e. dimensions, bulk density, and moisture content. The density was determined by means of test cubes manufactured from the concrete mixtures. A representative density of (1.89 ± 0.04) g/cm^3 for dry concrete

Table 2. Chemical compositions of the pads.

	% abundance					
	K-1	K-2	U-1	T-1	UT-1	UT-2
SiO_2	66.0	50.4	71.2	72.6	66.8	69.7
TiO_2	0.2	0.1	0.2	0.1	0.1	0.1
Al_2O_3	4.2	14.0	3.9	2.9	5.9	5.5
Fe_2O_3	0.9	0.9	0.8	0.7	2.7	2.0
MnO	0.0	0.0	0.0	0.0	0.1	0.0
MgO	0.5	0.4	0.4	0.4	0.3	0.3
CaO	19.0	17.2	15.0	15.4	15.1	14.7
Na_2O	1.0	2.0	0.8	0.5	1.7	1.3
K_2O	1.3	8.3	1.2	1.0	1.6	1.6
H_2O						
CO_2	6.9	6.7	6.5	6.4	5.7	4.8
Other						
	100.0	100.0	100.0	100.0	100.0	100.0

Table 3. Dimensions, density, and moisture content of the pads.

Diameter	300 cm
Thickness	50 cm
Volume	3.5 m^3
Bulk density with natural moisture	(2.01 ± 0.05) g/cm^3
Moisture content	6 ± 1 %

has been established. This value, corrected for a moisture content of (6 ± 1)% in the porespaces of the pads, furnishes a bulk pad density of (2.01 ± 0.05) g/cm^3. The moisture determination was performed by core drilling along the perimeters of several pads with the surrounding gravel partially removed.

3. PAD RADIOELEMENT CONCENTRATIONS

This section contains the details of efforts undertaken to establish a set of unbiased radiometric reference concentrations for use in instrument calibrations. Readers having no particular interest in this evaluation need only consult Table 6 in which the adopted pad radioelement concentrations are presented.

3.1. Laboratory assays

The manufacture of all six pads was accompanied by a continuous collection of grab-samples of mixture rammed down in the form (see Fig. 10). The material retrieved from the single pad mixtures amounted to several tens of kg. All samples were allowed to set and were then crushed and finely ground. This powdered material has been used for the preparation of six homogeneous

Fig. 10. Collection of grab-sample from wet mixture.

gross pad samples. Splits of these provided the bulk chemical compositions in Table 2. The radioelement assays performed are based on sealed-can NaI(Tl) gamma-ray spectrometry of six series of twenty 250-g splits.

The laboratory gamma-ray spectrometer operated by the Nuclear Geophysics Group is equipped with a horizontally mounted NaI(Tl) detector which has an active volume of 1287 cm^3 and is shielded by 10 cm of equivalent lead (Løvborg, 1972). A leadlined disk intersecting the front part of the shield serves as a feed-through device for the sample cans. These are made of sheet metal and hold 170 cm^3 of closely packed powder. The cans are filled up on a vibrating plate and are then sealed hermetically with lids imprented by rolling. A series of prepared samples is always stored for three weeks to await the buildup of equilibrium concentrations of radon gas in the cans. The actual recording of spectra is done with a multichannel analyser

that has been interfaced with the motor drive for the sample-changer wheel. The pad samples were dried for 24 hours at 110°C before they were sealed, and analysed by totalling the counts in spectral windows at 1.46 MeV (^{40}K), 1.76 MeV (^{214}Bi), and 2.62 MeV (^{208}Tl). The calibration used to determine the sensitivity factors and stripping ratios of the NaI(Tl) detector with associated energy windows was much more comprehensive than usual. The procedure involved the counting of several K salts plus a whole series of U and Th reference materials obtained from the New Brunswick Laboratory (NBL) in the U.S. U counting standards in strict radioactive equilibrium would have been assigned radiometric grades identical to the certified U concentrations. The NBL reference materials have been prepared from pitchblende with a ^{226}Ra surplus of 1.9% and were accordingly used as standards containing 1.019 ppm eU per ppm U.

Assayed radiometric pad concentrations, in units of % K, ppm eU, and ppm Th, are reported in Table 4. A column of elemental U concentrations, in units of ppm U, is included. These latter data are based on neutron irradiation and delayed-neutron counting of 1-g splits using equipment installed at Risø's research

Table 4. Laboratory assays of dry pad material. Values determined at Risø by gamma-ray spectrometry and delayed-neutron counting.

Pad	% K	ppm U	ppm eU	ppm Th
K-1	1.095 (0.007)	1.14 (0.03)	0.31 (0.04)	2.38 (0.09)
K-2	6.883 (0.027)	2.35 (0.06)	4.44 (0.10)	1.95 (0.13)
U-1	0.965 (0.057)	228.7 (9.7)	230.0 (3.6)	2.03 (0.50)
T-1	0.775 (0.058)	7.06 (0.18)	8.46 (0.54)	168.8 (1.04)
UT-1	1.404 (0.056)	57.19 (0.97)	59.90 (1.05)	155.6 (0.72)
UT-2	1.310 (0.025)	19.58 (0.35)	18.76 (0.42)	50.45 (0.71)

Numbers in parentheses are calculated standard deviations which include analytical accuracy and precision.

reactor DR 3 (Kunzendorf et al., 1980). All pads except K-1 and K-2 exhibit nearly the same eU and U concentrations and may be assumed to contain equilibrium amounts of ^{226}Ra. K-1 and K-2 appear to possess a smaller and larger ^{226}Ra content, respectively, than that corresponding to radioactive equilibrium. Several check assays were provided by counting laboratories in U.S.A., Australia, and South Africa among whom 1-kg splits of the gross pad samples had been distributed. The mutual agreement of these additional results plus the Risø determinations was acceptable though not impressive. The additional assays available for UT-2, which has been chosen as a representative example, are shown in Table 5.

Table 5. Check assays for pad UT-2. Data were provided by counting laboratories in three countries.

Laboratory	% K	ppm U	ppm eU	ppm Th
I	1.32	21.8	17.9	51.9
II	1.5	18.7	18.7	43.5
III	n.d.	17.6	18.8	61.9
Averages:	1.4 (0.1)	19.4 (1.3)	18.5 (0.3)	52.4 (5.3)

Numbers in parentheses are resulting standard deviations.

3.2. Adopted K-eU-Th reference concentrations

To obtain a set of reliable reference concentrations for the actual pads, the assay values in Table 4 were first divided by a factor of 1.06. This correction allows for an average moisture content of 6% in the pads. A further modification of the laboratory determinations was suggested by gamma-ray monitoring performed in situ. The experimental set-up is shown in Fig. 11. Pad window counts were recorded systematically from February 1980 to August 1981. In the same period the spectrometer was brought along on calibration trips to large pads installed at

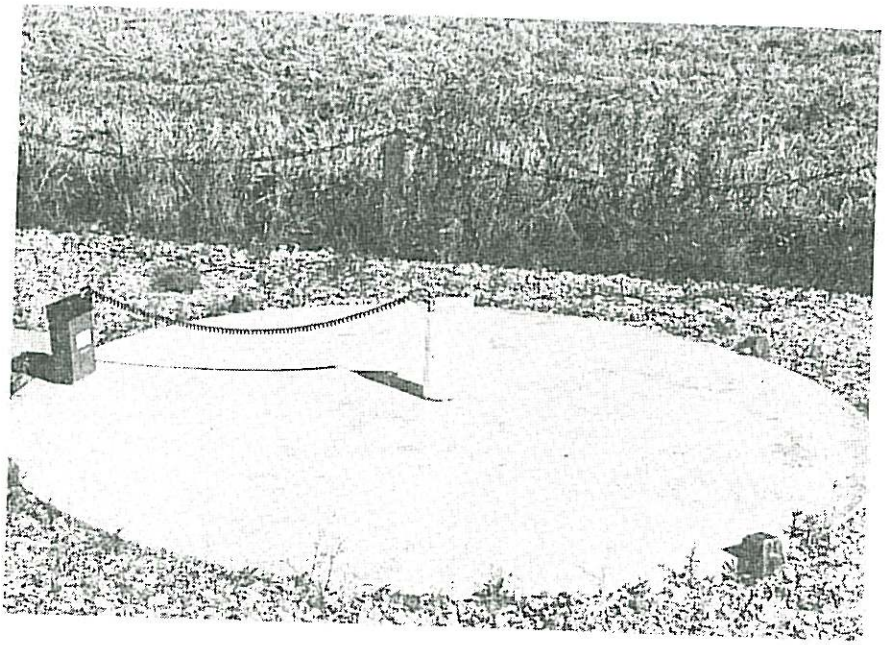

Fig. 11. Pad monitoring by means of a portable spectrometer (a Geometrics/Exploranium GR-410).

airports in U.S.A. (Grand Junction, Colorado), Canada (Ottawa), Finland (Helsinki), and Sweden (Borlänge). The calibration measurements provided the sensitivity factors and stripping ratios of the instrument, so that it became possible to convert the monitoring data for the Risø pads into an independent series of radioelement concentrations. Because the data contained an unknown and possibly varying background contribution, the least radioactive pad (K-1) served as a base for assigning a set of calculated difference concentrations to each of the other pads.

In Fig. 12, the laboratory K assays (moisture corrected) are plotted against the differential K concentrations evaluated from the monitoring experiment. The added regression line includes the known variances of both sets of observations. The vertical displacements of the experimental points with respect to the line represent the extra corrections imposed on the laboratory determinations. K pad concentrations derived in this way are smoothed values that vary in close accordance with the ^{40}K gamma radiation detected on the pads. Since the procedure involves no change of scale, the results are unaffected by the

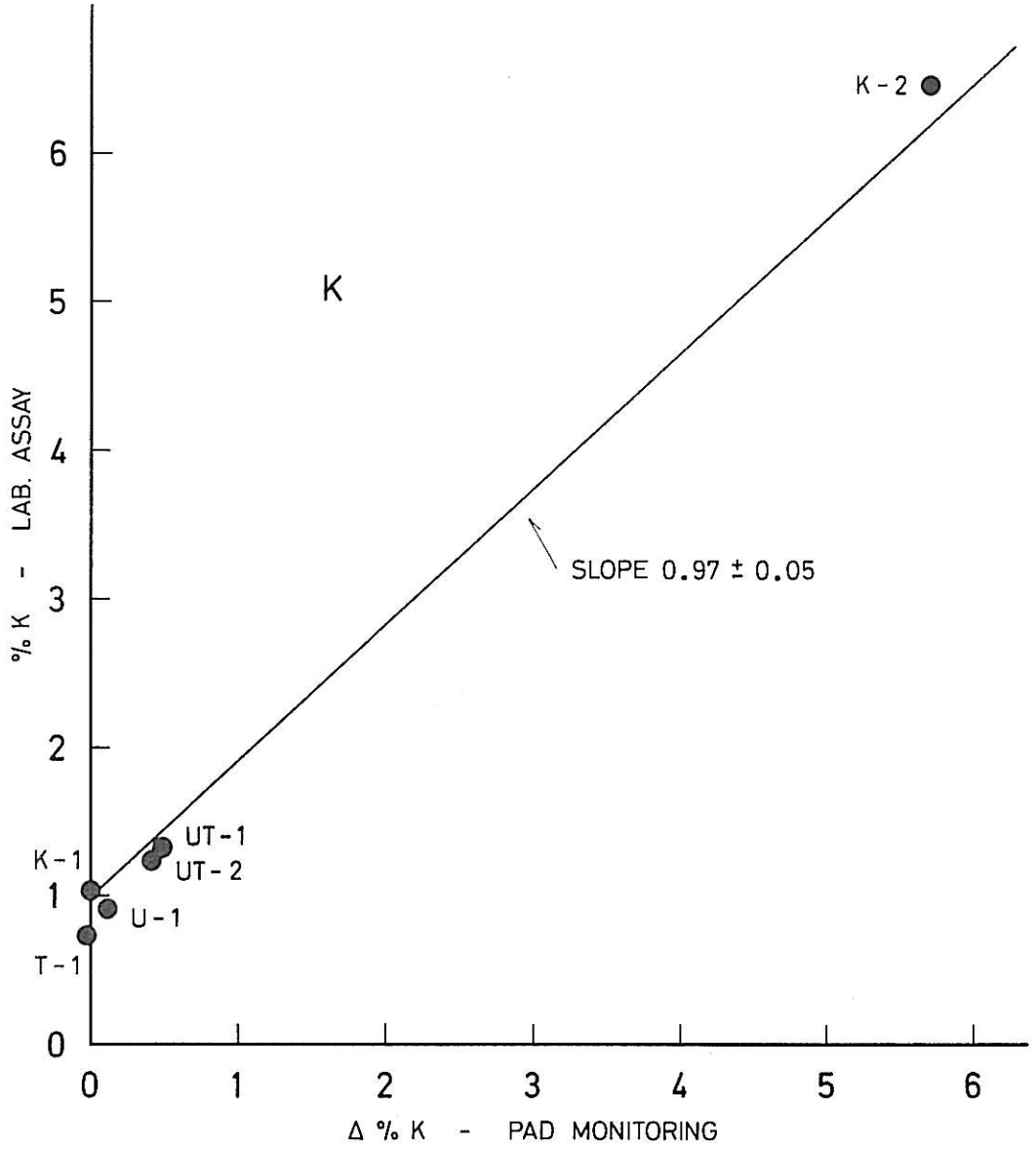

Fig. 12. Experimental potassium concentrations for the pads. Concentration differences are with respect to pad K-1.

accuracies of K concentrations in the airport pads used for calibration. Figures 13 and 14 illustrate the corresponding data from which the final pad concentrations of eU and Th have been established. The slope of the eU regression line is significantly greater than unity and has a rather wide confidence interval. In view of the extraordinary means applied to ensure the accuracy of the laboratory assays, it is unlikely that analytical error is the only reason for the systematic deviation from a 1:1 correspondence. The problem may be solved after a

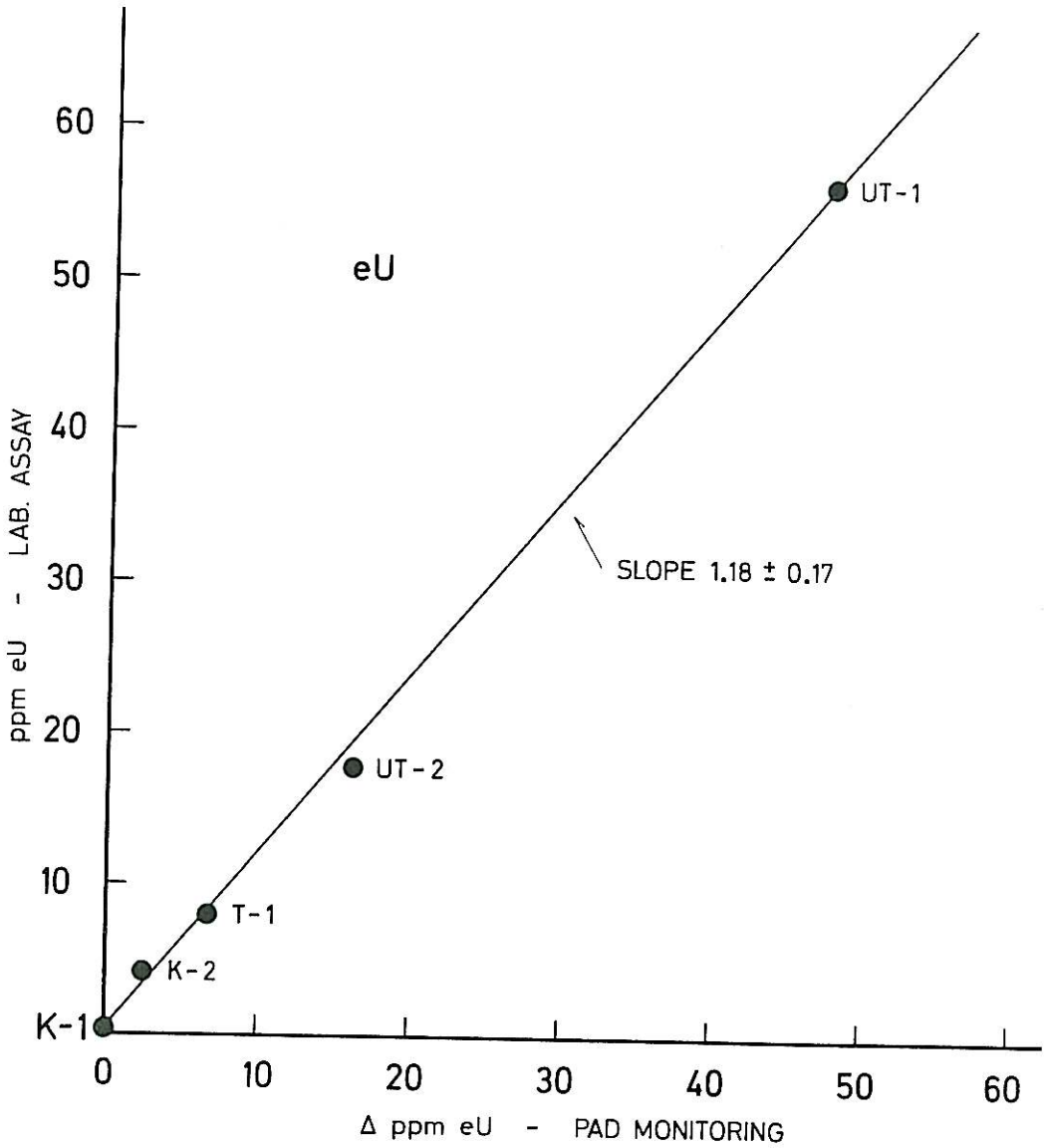

Fig. 13. Experimental uranium concentrations for the pads. Concentration differences are with respect to pad K-1.

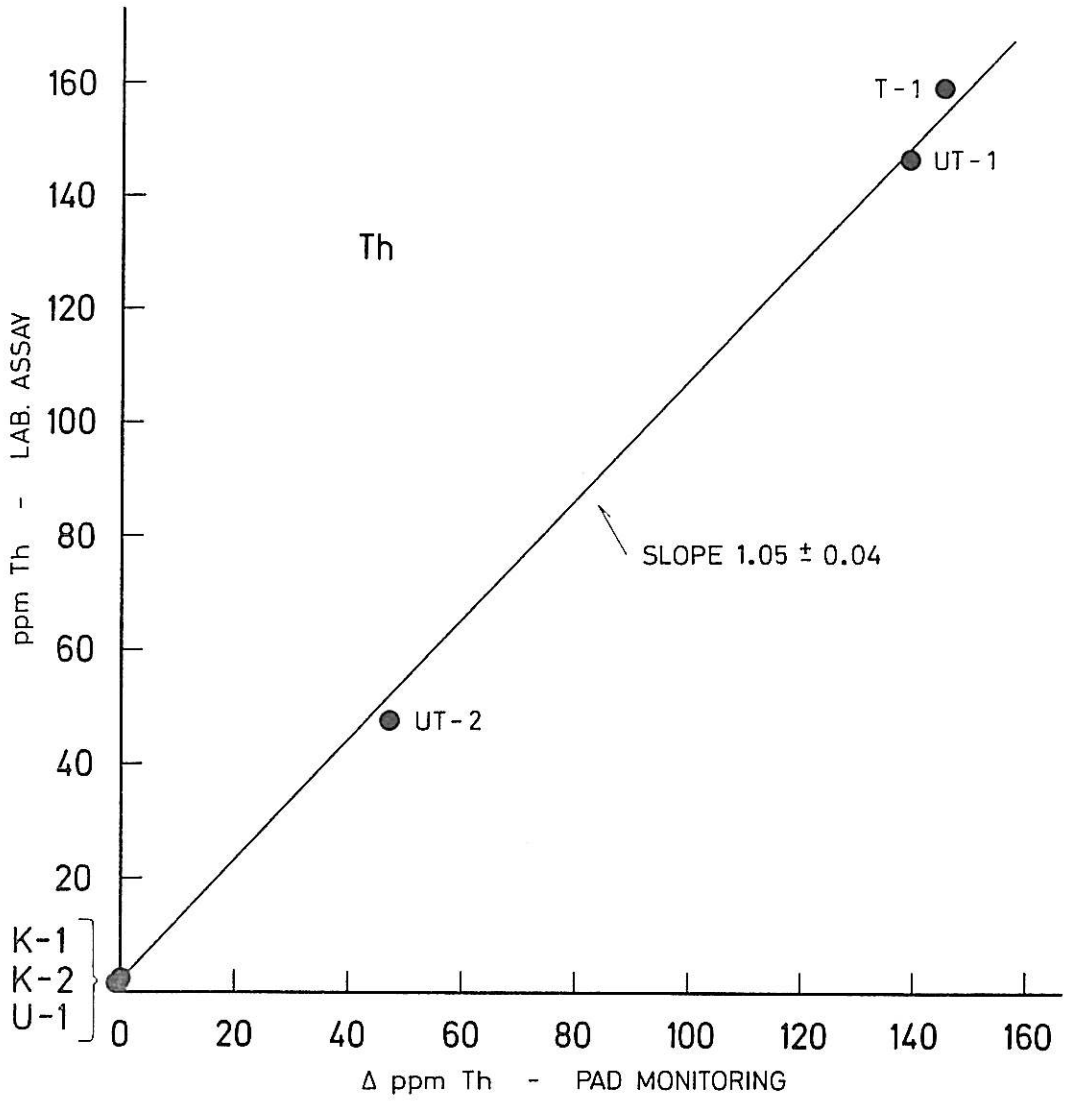

Fig. 14. Experimental thorium concentrations for the pads. Concentration differences are with respect to pad K-1.

careful re-examination of the U sensitivity factors recorded with the individual airport pad facilities.

Pad U-1 has been excluded from the regression in Fig. 13 and Fig. 15 explains why. This additional plot shows the series of eU concentrations determined by pad monitoring in 1980 after a normalization to equivalent laboratory determinations. The records for UT-1 and UT-2 suggest that these pads retain their equilibrium concentrations of gamma-ray emitters all year round. U-1, on the other hand, appears to lose a substantial fraction of its activity between March and November. This

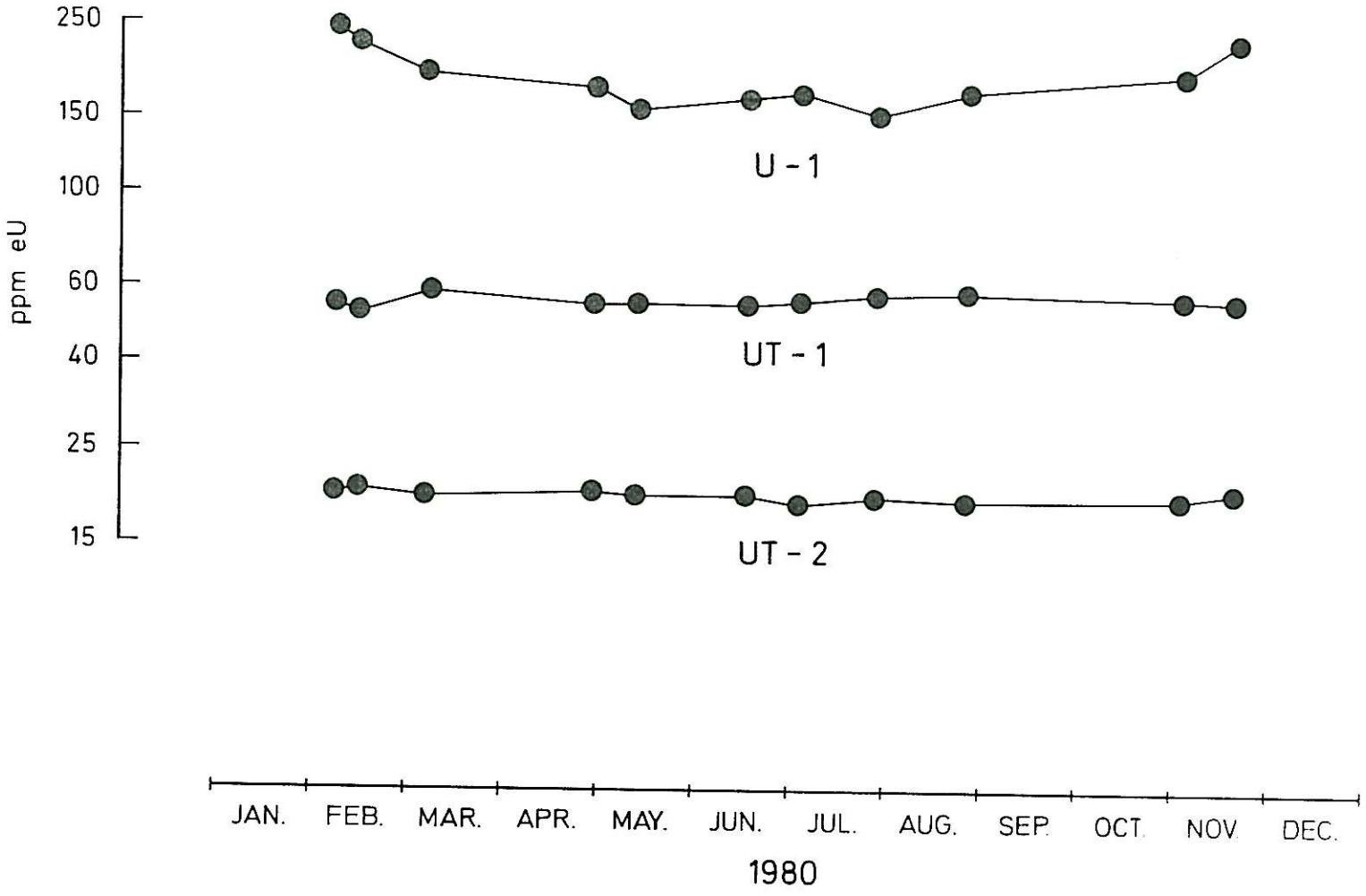

Fig. 15. Monitoring results for pads loaded with uranium.

seasonal variation of the eU concentration has been observed for almost a decade and is ascribed to a varying outflow of radon from the pad (Løvborg et al., 1978). In the winter the pad is effectively sealed off by remanent surface moisture or ice and should attain an equilibrium concentration of 217 ppm eU (230/1.06, cf. Table 4). This level has actually been exceeded many times (see the observations for February and late November in Fig. 15). The high-grade uranium ore added to the pad was perhaps not properly mixed up with the concrete and may be distributed with an excessive abundance near the top surface.

The adopted set of radiometric reference concentrations is presented in Table 6. New values ascribed to the old pads K-1, K-2, and T-1 do not differ much from assays produced in the past (Løvborg and Kirkegaard, 1974; Løvborg et al., 1978). U-1

Table 6. Adopted reference concentrations for use in instrument calibrations on the pads.

Pad	% K	ppm eU	ppm Th
K-1	0.995 (0.041)	0.39 (0.21)	2.07 (0.29)
K-2	6.475 (0.145)	3.28 (0.31)	2.14 (0.30)
U-1	1.107 (0.083)	170 (25)	1.57 (0.62)
T-1	0.970 (0.047)	8.28 (0.65)	154.6 (3.2)
UT-1	1.469 (0.053)	56.5 (4.2)	148.4 (3.2)
UT-2	1.386 (0.044)	19.50 (1.45)	51.9 (1.1)

Numbers in parantheses are estimated standard deviations.

is now described by an eU concentration that is representative of the reduced radon content in this particular pad during the period in which most instrument calibrations are executed. The reported standard deviations are believed to be quite realistic. They include the combined variance of the laboratory assays and the averaged monitoring results used to correct the latter. Fur-

thermore the standard deviation of the eU concentration assigned to U-1 incorporates a variance associated with the seasonal variation of the emitted gamma radiation.

4. RECORDING AND PROCESSING OF CALIBRATION COUNTS

The majority of field instruments received for calibration at Risø are fully portable and can easily be centred on the pads in direct contact with them. It is recommended practice to record calibration counts over several consecutive counting intervals. The readings obtained on the single pads are written down on standard forms and are then combined into single data sets. The numerical evaluation of calibration constants is performed with assistance from a terminal connected to the Burroughs central computer at Risø (presently a B-6700, which will be replaced by a B-7800 in 1982). Total-count sensitivity factors are provided by executing the program PADSUM. Spectral window sensitivities and stripping ratios are calculated by means of PADWIN. Both programs are based on the same mathematical approach and numerical technique. (Readers interested in the data reduction method will find the relevant details in subsection 4.3.)

4.1. Calibration of total-count scintillometers

Figure 16 exemplifies the calibration of a field scintillometer of a type familiar to many uranium geologists in Europe. This particular instrument displays the instantaneous count rate on a moving-coil meter, and a single reading corresponds to an effective counting time of 2τ, where τ (= RC) is the integration constant used (Price, 1958). A filled-in standard form with a series of ratemeter readings and their equivalent counting data is presented in Fig. 17. The resulting measuring time and number of counts recorded on each pad are keyed in as input to the

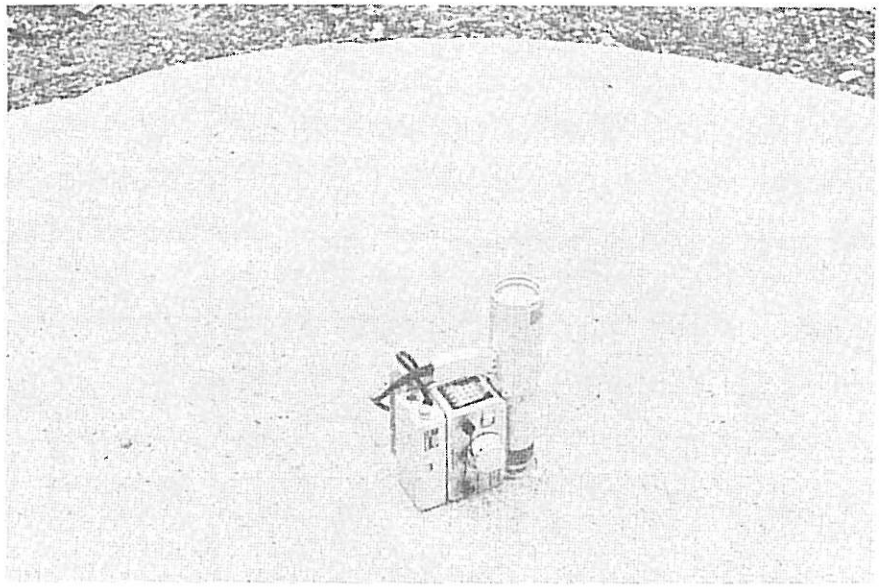

Fig. 16. Pad calibration of a total-count scintillometer (a Saphymo S.P.P.2 NF).

PADSUM program. The latter prints the most likely values and the standard deviations of the count rates produced by 1% K, 1 ppm eU, and 1 ppm Th. It has been found that pad U-1 with its poorly defined eU concentration does not improve the U sensitivities estimated with pads UT-1 and UT-2 alone. U-1 is therefore left out in the recording of total-count calibration data.

The U sensitivity is the calibration constant of actual interest to the user of a total-count scintillometer. This has background in a generally accepted recommendation according to which total gamma-ray signals should be reported as apparent U concentrations (IAEA, 1976; IAEA, 1979). A division of the U sensitivity into the count rate produced by outcropping rock furnishes the total radioactivity of the outcrop in units of radioelement concentration, abbreviation Ur *). Ur values measured on pure uranium mineralization are identical to the

*) The symbol Ur was suggested in 1980 by the OECD Nuclear Energy Agency. It replaces the symbol introduced by IAEA (1976), ur, which might be confused with microröntgens.

Calibration of Saphymo S.P.P.2 NF, serial 2064. 1981-05-04.

PAD	RECORDED SIGNAL	EQV. COUNTING TIME (S)	EQV. NUMBER OF COUNTS
UT-1			
	600 cps	0.8	480
	550 -	0.8	440
	500 -	0.8	440
		2.4	1360
UT-2			
	280 cps	1.2	336
	290 -	1.2	348
	290 -	1.2	348
		3.6	1032
K-1			
	120 cps	8.0	960
	120 -	8.0	960
	120 -	8.0	960
		24.0	2880
K-2			
	180 cps	1.2	216
	160 -	1.2	192
	170 -	1.2	204
		3.6	612
T-1			
	420 cps	1.2	504
	390 -	1.2	468
	390 -	1.2	468
		3.6	1440

Fig. 17. Recorded total-count calibration data.

readings in ppm eU that would have been obtained with a portable spectrometer. In all other cases Ur determinations express the combined concentration of K, eU, and Th in the ground.

Table 7 summarizes total-count calibration constants determined on the Risø pads by users of four different types of portable scintillation counters. The third column shows the experimental U sensitivities stated as generally applicable total-count sensitivities in units of counts/s per Ur. The use of count rate

Table 7. Experimental total-count sensitivities for portable scintillation counters.

Instrument	Number of instruments calibrated	Counts/s per Ur	Ur per ppm Th	Ur per % K
Saphymo S.P.P.2 NF	8	3.68(0.39)	0.425(0.048)	1.53(0.50)
Scintrex BGS-2	5	4.83(0.48)	0.463(0.065)	1.59(0.33)
Scintrex BGS-3	2	8.60(0.79)	0.436(0.011)	1.69(0.25)
McPhar TC 33A	2	4.89(0.28)	0.459(0.029)	1.95(0.22)

Numbers in parentheses are estimated standard deviations per single instrument.

per Ur is justified by the numbers in the two additional columns on the right. These show the experimental Th and K sensitivities in units of Ur per ppm Th and % K. Th and K sensitivities expressed in this way are seen to vary very little from one instrument to another, and this is why it is meaningful to characterize a counter response by just one figure: the count rate produced by one Ur. Løvborg et al. (1976) found that gross scintillation counting theoretically should provide responses of 0.44 Ur per ppm Th and 1.6 Ur per % K. The calibration data in Table 7 result in estimates of (0.438 ± 0.013) Ur per ppm Th and (1.70 ± 0.18) Ur per % K. The good agreement between predicted and observed values may be taken as evidence that the ratios between the radioelement concentrations of the pads are fairly reliable.

4.2. Calibration of portable gamma-ray spectrometers

The recording of spectrometer window counts proceeds as illustrated for the monitoring experiment in Fig. 11. The instrument shown accumulates counts for a preset time interval and presents the window contents in succession on a four-digit luminescent display. Repeated pad counting is accordingly necessary to obtain a minimum of 10,000 counts without overflow. A standard form with a complete set of calibration data is exemplified in Fig. 18. The K counts (1.46 MeV), U counts (1.76 MeV), and Th counts (2.62 MeV) are inputs to the PADWIN program by means of which the following calibration constants are estimated:

Sensitivity factors:

s_K = K count rate per % K
s_U = U count rate per ppm eU
s_T = Th count rate per ppm Th .

Stripping ratios:

α = U counts per Th count for Th radiation
β = K counts per Th count for Th radiation
γ = K counts per U count for U radiation
a = Th counts per U count for U radiation .

It may be usefull to recall the equations in which these constants are used to process window count rates n_K, n_U, n_T produced by outcropping rock:

Stripping equations:

$$\begin{aligned} n'_T &= (n_T - a\, n_U)/(1 - a\alpha) \\ n'_U &= (n_U - \alpha\, n_T)/(1 - a\alpha) \\ n'_K &= n_K - \beta\, n'_T - \gamma n'_U \quad . \end{aligned} \tag{1}$$

Calibration of Geometrics GR-410, serial 3041. 1981-10-02.

PAD	TIME (S)	WINDOW COUNTS			
		TOTAL	K	U	Th
UT-1					
	120	64530	2842	2896	1996
	120	64440	2731	2711	1994
	120	64290	2888	2767	2035
	360	193260	8461	8374	6025
UT-2					
	240	52570	2658	2078	1400
	240	53180	2649	2087	1446
	240	52780	2648	2097	1426
	720	158530	7955	6262	4272
K-1					
	600	18710	1123	280	139
	600	19060	1124	326	148
	600	18830	1057	329	155
	1800	56600	3304	935	442
K-2					
	240	31550	4560	370	85
	240	31890	4565	384	95
	240	31550	4525	392	105
	720	94990	13650	1146	285
T-1					
	240	86770	3090	2925	4230
	240	86580	3067	2881	4198
	240	87370	3101	2970	4231
	720	260720	9258	8776	12659
U-1					
	60	44130	2365	2633	83
	60	44640	2394	2608	87
	60	44290	2377	2689	88
	180	133060	7136	7930	258

Fig. 18. Pad window counts recorded with a portable spectrometer.

Conversion equations:

$$\begin{aligned} \%\ K &= n'_K/s_K \\ \text{ppm eU} &= n'_U/s_U \\ \text{ppm Th} &= n'_T/s_T \quad . \end{aligned} \tag{2}$$

Sensitivity factors and stripping ratios for 4 identical spectrometers with consecutive serial numbers are shown in Tables 8 and 9. In this real calibration example it appears that the K window of instrument 3043 was slightly out of adjustment. This shows that the pads are very useful for checking out newly purchased equipment. The small values of the stripping ratio, a, are typical of NaI(Tl) detectors with good energy resolution. This calibration constant is the only one that cannot be reliably determined without including pad U-1 in the calibration measurements. Its incorporation in the stripping equations may not be necessary in some field applications, but is essential for the determination of Th in uranium ore.

Table 8. Experimental window sensitivities for 4 Geometrics/Exploranium (model GR-410) portable spectrometers.

Serial number	s_K counts/s per % K	s_U counts/s per ppm eU	s_T counts/s per ppm Th
3041	2.959(0.086)	0.254(0.016)	0.1124(0.0016)
3042	3.288(0.095)	0.255(0.016)	0.1131(0.0016)
3043	2.437(0.072)	0.260(0.016)	0.1142(0.0016)
3044	3.148(0.091)	0.252(0.016)	0.1103(0.0016)

Numbers in parentheses are estimated standard deviations.

The total-count window of a spectrometer usually has a lower energy limit of several hundred keV. The recorded signal is not the gross gamma-ray intensity measured with the simple scintillation counters whose calibration constants were discussed in

Table 9. Experimental stripping ratios for 4 Geometrics/ Exploranium (model GR-410) portable spectrometers.

Serial number	α	β	γ	a
3041	0.566(0.018)	0.544(0.020)	0.900(0.025)	0.025(0.004)
3042	0.554(0.018)	0.482(0.020)	0.835(0.025)	0.028(0.004)
3043	0.544(0.018)	0.466(0.019)	0.893(0.021)	0.026(0.004)
3044	0.562(0.018)	0.513(0.020)	0.802(0.024)	0.028(0.004)

Numbers in parentheses are estimated standard deviations.

the preceding subsection. Løvborg et al. (1976) demonstrated that Th and K sensitivities expressed in Ur per concentration unit of Th and K vary appreciably with the energy threshold. For this reason the calibration practice at Risø does not include the determination of spectrometer total-count sensitivities in units of counts/s per Ur. Instead it may be possible to supply a reliable factor for converting total count rates into environmental exposure rates (subsection 5.2).

4.3. Use of nonlinear statistical techniques

PADSUM and PADWIN are programs which make it possible to perform instrument calibration with a series of more than four pads. The least squares techniques used are conveniently described with the assistance of two index numbers, i and j. The first of these selects a particular pad and the other identifies the radioelements in the succession K, U, and Th. A set of pad count rates recorded totally or in a spectrometer window is then represented by the equations:

$$n_i = \sum_{j=1}^{3} a_j x_{ij} + b \quad , \tag{3}$$

where

a_j = the unknown count rate produced by a unit concentration of the j´th radioelement;

x_{ij} = the concentration of the j´th radioelement in the i´th pad;

b = an unknown background.

(3) fits the general linear model $\underline{n} = X\,\underline{a}$ in which $\underline{n}$ is a vector of k independent observations, $\underline{a}$ a vector of m unknowns, and X a known matrix with k rows and m columns. For calibrations on the Risø pads, k is either 5 or 6, depending on whether or not it is relevant to include pad U-1 in the data recording. The actual value of m is 4, and $\underline{a} = (a_1, a_2, a_3, b)$ is obtained by minimizing the weighted sum of squared residuals:

$$\sum_{i=1}^{k} \left[\frac{r_i(\underline{a})}{s_i(\underline{a})}\right]^2 = \text{minimum} \tag{4}$$

with

$$r_i = n_i - \sum_{j=1}^{4} a_j x_{ij} \tag{5}$$

$$s_i^2 = \mathrm{Var}[n_i] + \sum_{j=1}^{4} a_j^2 \,\mathrm{Var}[x_{ij}] \tag{6}$$

$$x_{i4} = 1 \tag{7}$$

$$\mathrm{Var}[x_{i4}] = 0 \quad . \tag{8}$$

In the variance expression (6), $\mathrm{Var}[n_i]$ is given by the number of counts recorded on the i´th pad. $\mathrm{Var}[x_{ij}]$ (j<4) are the squared standard deviations of the pad radioelement concentra-

tions (Table 6). Because the variance supplied by the pad concentrations depends on $\underline{a}$ through the factors a_j^2, the regression problem presented by (4) is nonlinear and must be solved by iteration. The iteration strategy selected is that contained in the Levenberg-Marquardt optimization code LMDER1 from the MINPACK-1 collection developed at the U.S. Argonne National Laboratory by Moré et al. (1980). The iterations are started from a first estimate of $\underline{a}$ provided by an unweighted least-squares fit. An expression for evaluating the variances and covariances associated with a final solution vector has been established from a first-order perturbation analysis of the problem (4).

The difference between PADSUM and PADWIN lies in the stripping ratio calculation executed by the latter. The immediate result of the iterations performed by PADWIN is a set of three $\underline{a}$ vectors which may be combined into a spectrometer response matrix, A. An element of A, a_{wj}, is then the count rate produced by a unit concentration of the j'th radioelement in the w'th spectrometer window. Stripping ratios are calculated as

$$p/q = a_{wj}/a_{jj} \qquad (w \neq j) \quad . \tag{9}$$

It can be shown that

$$\mathrm{Var}[p/q] \approx q^{-2}\,\mathrm{Var}[p] - 2pq^{-3}\,\mathrm{Cov}[p,q] + p^2q^{-4}\,\mathrm{Var}[q] \quad . \tag{10}$$

Cov[p,q], the covariance of counts from the same radioelement in different windows, is derivable from an extension of the perturbation analysis of an $\underline{a}$ vector. The estimated standard deviations of spectrometer calibration constants typically amount to

3 to 5% for K window sensitivities
6 to 8% for U window sensitivities
2 to 4% for Th window sensitivities
3 to 4% for the stripping ratios α, β, and γ.

Actually calculated standard deviations have been verified using repeated PADWIN runs in which the pad concentrations and the window counts fluctuated randomly in accordance with their known variances.

5. SUPPLEMENTARY INFORMATION

5.1. Field application of pad calibrations

Counters and spectrometers are used in many different geologic environments that involve measuring conditions only exceptionally identical to those realized with pads. Assays of thin veins and other occurrences of small surface extension appear to present a calibration problem inconsistent with the possibilities offered by the facility promoted here. An increasing number of geologists have experienced that it is difficult anyhow to obtain reliable grade estimates on mineralization that is not uniformly distributed within the viewing radius of the gamma-ray detector. In the U.S. it is common practice to perform total-count assays from readings taken with and without a small filter plate inserted between the rock face and the counter (Dodd, 1976). These differential scanners detect the radiation from a small volume of rock and furnish quantitative concentration data when calibrated on pads. The technique efficiently removes many of the problems associated with in-situ assays and deserves attention among uranium geologists in Europe.

A reverse and possibly less common situation is that encountered in assays of whole-rock radioelement concentrations in large outcrops. Measurements like these favour the use of a large effective sample and are conveniently executed in the natural geometry of nearly 2π. Instrumental sensitivity factors recorded with the pads at Risø are slightly smaller than those required on outcrops larger than 10 m^2 in area. Provided

the outcrop diameter can be estimated with some confidence, it may be possible to correct a set of experimental sensitivity factors. The corrections are performed by running the gamma-ray transport code GAMO which the authors developed as a means of estimating instrument responses produced by cylindrical sources. GAMO simulates gamma-ray transport by scattering and absorption using Monte Carlo techniques described by Kalos (1968). The code gets the necessary K-U-Th emission spectra, gamma-ray interaction cross sections, and detector response functions from a system of data files which serve a variety of simulation tasks at Risø (Løvborg and Kirkegaard, 1974; Løvborg et al., 1976; Kirkegaard and Løvborg, 1979; Kirkegaard and Løvborg, 1980).

Figure 19 shows the calculated effect of the outcrop diameter in gross-count and spectral field assays carried out with the NaI (Tl) detector placed directly on the ground. These data are normalized to a common sensitivity of unity in the preceding instrument calibration on the pads. Spectrometer counting may be accommodated to the geometry on an outcrop through a minor adjustment of the window sensitivities. Exposed rock faces that are more than 20 m across are effectively infinite sources with an associated correction factor of 1.06 for sensitivity factors recorded at Risø. Predicted relative sensitivities were used in reverse order when the pads were monitored for K-U-Th concentrations with a spectrometer that had been calibrated on large airport pads (subsection 3.2). The gross-count data presented in Fig. 19 demonstrate that it may be rather difficult to asses the actual sensitivity of measurement provided by an ordinary field scintillometer. A small NaI(Tl) crystal is an efficient detector of low-energy radiation, and typically 80% of the counts above 50 keV are produced by gamma-rays that have been energy-degraded by scattering in the ground and the air. The contribution from scattered radiation is an increasing function of the source diameter up to a saturation point that is never reached on outcrops of usual dimensions. The gross-count curve in Fig. 19 may be useful for estimating scintillometer sensitivities on rock faces that are between 3 and 20 m in diameter. Because the curve is steep and valid for flat ground only, it is not applicable for correction purposes in assays of larger exploration targets.

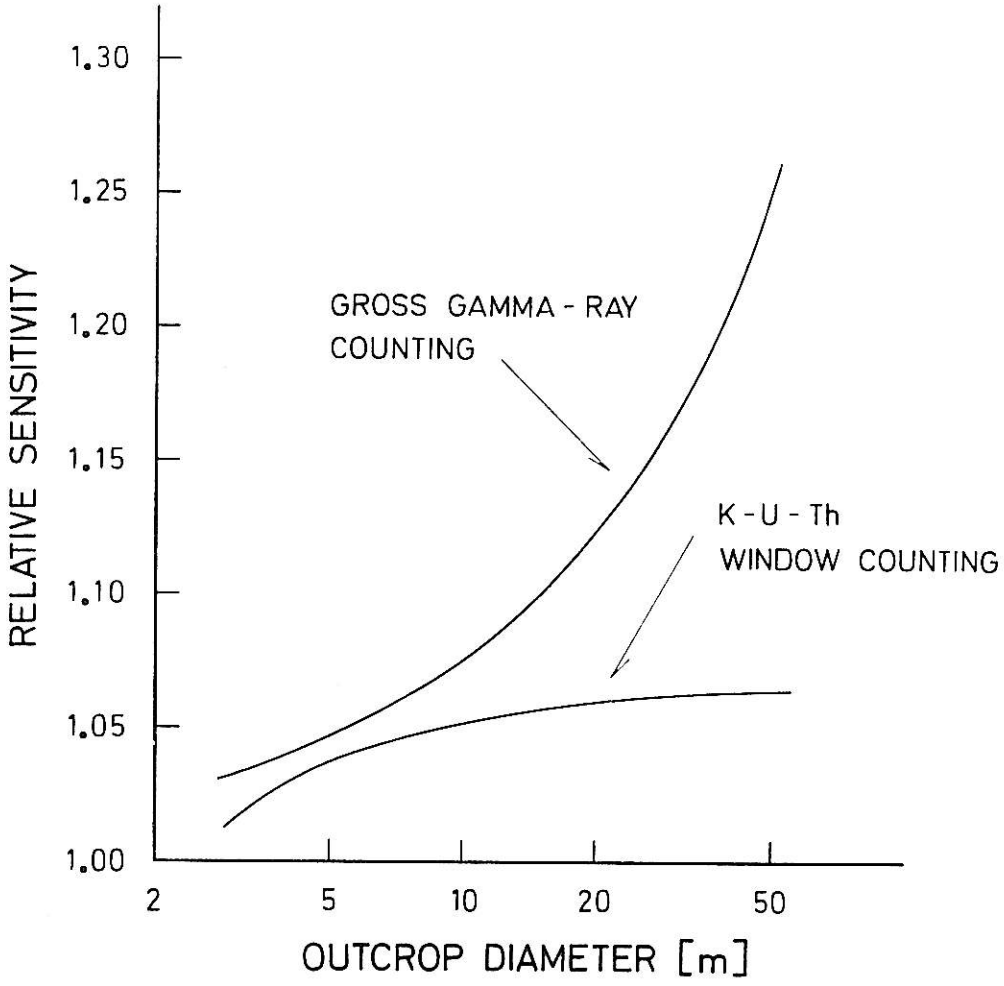

Fig. 19. Calculated responses of counters and spectrometers on 10-m thick outcropping rock.

5.2. Exposure rates produced by the pads

The gamma radiation contributed by a calibration pad is spatially uniform and has the spectral characteristics of a natural radiation environment. A facility like that presented here may therefore serve calibration tasks implicated by environmental radiation monitoring. The gamma-ray exposure rates produced by all pads except the unreliable U-1 have been determined with good accuracy, and the facility is henceforward established as a supplementary reference target for radiation surveys undertaken by Risø.

Total exposure rates assigned to the pads are presented in Table 10. The data include four recordings with a Reuter-Stokes

Table 10. Total exposure rates evaluated from monitoring with ionization chamber and TL dosimeters.

Pad	μR/h
K-1	9.78 (0.61)
K-2	17.36 (0.66)
U-1	-
T-1	44.07 (0.38)
UT-1	62.61 (0.51)
UT-2	27.90 (0.32)
Background	8.05 (0.62)

Numbers in parentheses are estimated standard deviations which reflect the precision of repeated single measurements.

spherical ionization chamber plus dose integrations with two sets of LiF TL dosimeters over a period of six weeks. Monitoring with the ionization chamber is illustrated in Fig. 20. Calibration of the ionization chamber in units of μR/h is based on outdoor measurements on a certified 1-mCi ^{226}Ra source with due consideration of background and ground-scattered source gamma-rays (Nielsen and Bøtter-Jensen, 1981). The TL dosimeter readings were calibrated using ^{60}Co reference doses supplied with a TL irradiation facility available at Risø (Bøtter-Jensen and Nielsen, 1981).

Table 11 shows experimental and calculated exposure rates from unit concentrations of K, eU, and Th in the pads. The experimental values were obtained by running the PADSUM program with the ionization chamber data and the TL results treated as statistically equivalent counts. This processing furnished an additional estimate of the background radiation on the pads (see Table 10, bottom). The background is contributed by cosmic rays and scattered gamma-rays from the area surrounding the pads in

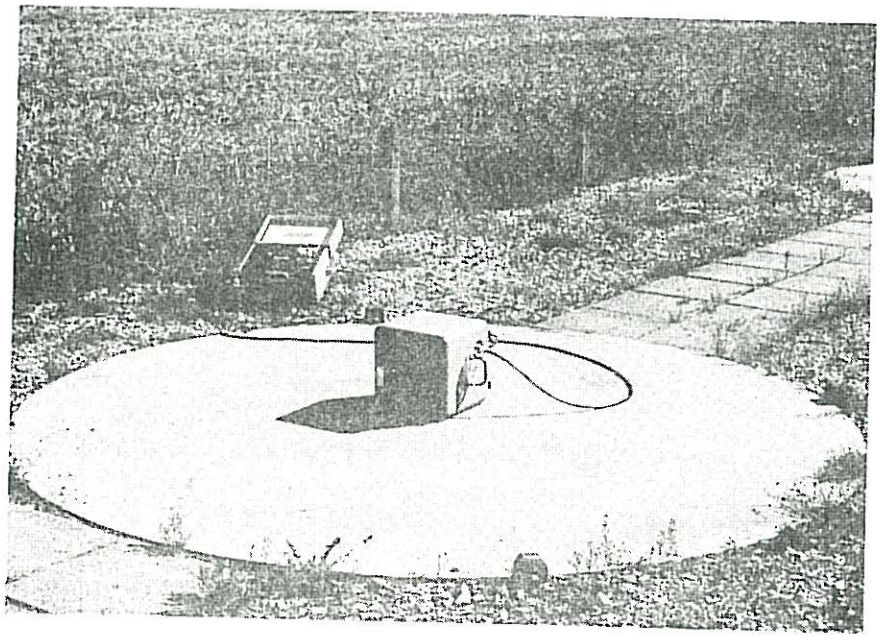

Fig. 20. Pad monitoring with a Reuter-Stokes model RSS-111 ionization chamber system.

Table 11. Estimated pad exposure rates per concentration unit of the radioelements in the pads.

	μR/h		
	1 % K	1 ppm eU	1 ppm Th
Experimental	1.17(0.06)	0.398(0.031)	0.204(0.009)
Calculated	1.22(0.01)	0.483(0.005)	0.240(0.002)
Ratio	0.96(0.05)	0.82 (0.06)	0.85 (0.04)

Numbers in parentheses are estimated standard deviations.

almost equal amounts. The calculated exposure rates in Table 11 were provided by the GAMO code. It can be seen that potassium is the only radioelement for which agreement between experiment and calculation is satisfactory. The experimental U and Th exposure rates are each approximately 15% smaller than their cal-

culated counterparts. This discrepancy is believed to be associated with the low-energy gamma-rays emitted by U and Th daughters. The pad surfaces are not perfectly smooth and plane, so that the escape probability for radiation of low energy may be smaller than that implied by the model assumption.

The three experimental figures in Table 11 have been combined with the pad concentrations in Table 6 for estimating the exposure rates contributed by the single radioelements in the pads. The resulting set of supplementary reference data for the pads is given in Table 12. They are used in conjunction with PADSUM in determining instrument responses produced by unit ex-

Table 12. Adopted gamma-ray exposure rates for use in instrument calibrations on the pads.

	μR/h		
Pad	K radiation	U radiation	Th radiation
K-1	1.16 (0.05)	0.16 (0.08)	0.42 (0.06)
K-2	7.54 (0.21)	1.30 (0.13)	0.44 (0.06)
T-1	1.13 (0.06)	3.29 (0.27)	31.51 (0.67)
UT-1	1.71 (0.07)	22.5 (1.7)	30.24 (0.67)
UT-2	1.62 (0.06)	7.76 (0.59)	10.58 (0.23)

Numbers in parentheses are estimated standard deviations.

posure rates of K, U, and Th radiation. An application is demonstrated in Fig. 21, which shows the result of calibrating the total-count response of a 76 x 76 mm NaI(Tl) detector in units of μR/h. Løvborg and Kirkegaard (1974) presented similar theoretical curves suggesting that 0.37 MeV would be an optimum energy threshold, providing nearly the same count rate per μR/h of K,U, and Th exposure. The uncertainty of the data in Fig. 21 does not make it possible to delimit a specific optimum thres-

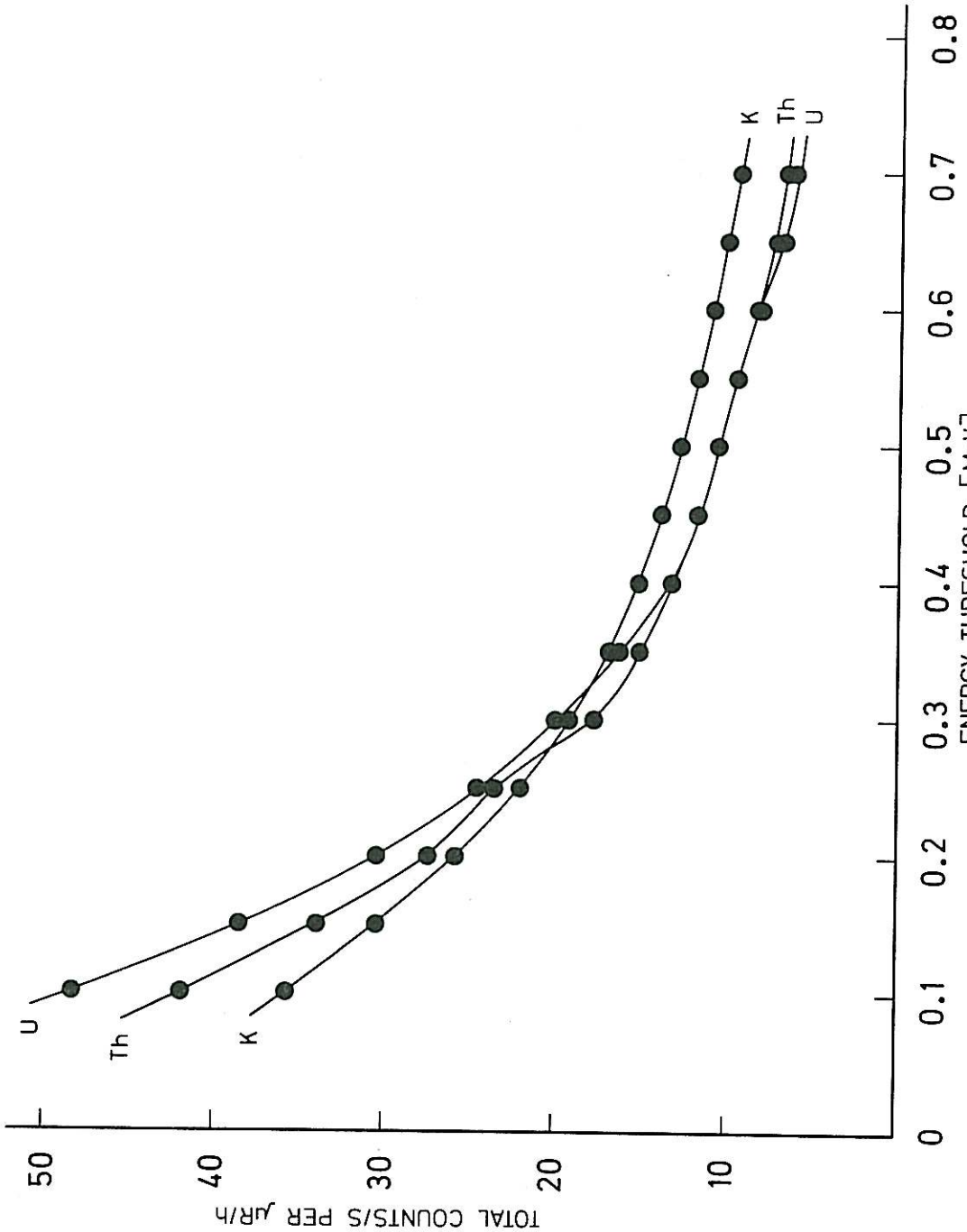

Fig. 21. Experimental total-count response of 76 x 76 mm NaI (Tl) detector to unit exposure rates of K, U, and Th radiation.

hold, but the experimental evidence clearly favours an energy cut-off of betwen 0.25 and 0.40 MeV for the measurement of environmental exposure rates by total gamma-ray counting. The total-count window of a portable spectrometer may be used for reasonably accurate exposure rate determinations, even for the window limit of 0.5 MeV necessitated by a detector with an installed ^{133}Ba energy-stabilization source (Løvborg et al., 1979). Simple field scintillometers with their low thresholds of 50 to 100 keV underestimate the exposure rate contributions from K and Th and should invariably be calibrated in Ur rather than in μR/h.

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